Layered rhyolite bands in a thick North Mountain Basalt flow: the products of silicate liquid immiscibility?

JOHN D. GREENOUGH*

Department of Geology, Mount Allison University, Sackville, New Brunswick, E0A 3C0

AND

J. DOSTAL

Department of Geology, St. Mary's University, Halifax, Nova Scotia, Canada, B3H 3C3

Abstract

The upper 35 m of a thick (≤175 m) Early Jurassic North Mountain Basalt flow at KcKay Head contains 25 cm thick differentiated layers that are separated by 130 cm sections of basalt. The lower layers are mafic, pegmatitic, and contain thin (2 cm), fine-grained 'rhyolite' bands. Evidence that the rhyolite represents a Si-rich immiscible liquid includes: (1) textures such as fine-grained globules of Fe-rich pyroxene (once Fe-rich liquid) bordering pegmatite feldspar grains; (2) structureless, microcrystalline, interstitial, polygonal patches of Si-rich minerals and similar areas of Fe-rich stilpnomelane surrounding skeletal Fe–Ti oxide grains, with bulk chemical compositions (to a first approximation), relative proportions and total modal percentages suggesting they were once Si-rich and Fe-rich glasses respectively; (3) basalt and pegmatite compositions (particularly their Fe, and Ti contents) similar to rocks known to contain immiscible liquids; (4) rhyolite major element compositions generally consistent with formation from an immiscible Si-rich liquid; (5) mineral compositions and temperature of pegmatite formation compatible with immiscibility; (6) the inability of mass balance calculations (crystal fractionation) to explain rhyolite formation unless mesostasis stilpnomelane (representing the Fe-rich liquid) is included in the caculations. If, as we suggest, these rocks are the result of immiscibility, they shed light on the incipient formation of granophyres in mafic intrusions and support liquid immiscibility as an important rock-forming process.

KEYWORDS: rhyolite, immiscibility, silicate liquid, North Mountain Basalt.

Introduction

FOLLOWING Bowen's (1928) conclusion that liquid immiscibility is not an important process in the differentiation of mafic rocks, most petrologists concentrated on documenting the effects of crystal fractionation on magma evolution. In 1970 Roedder and Weiblen reported that many lunar rocks contain globules of glass within a second chemically distinct glass. Since then numerous examples of immiscibility have been reported for terrestrial basalts (Roedder and Weiblen, 1971; De, 1974; Philpotts, 1976, 1978, 1979, 1982). This work has demonstrated that many basalts, in particular tholeiites, pass through a two-liquid stage during crystallisation. However the irrefutable evidence for immiscibility, one glass inside another is lost if the liquids separate. Thus the critical question, whether immiscibility is an important rock-forming process or is restricted to the mesostasis of basaltic rocks, remains unanswered. Although the importance of immiscibility remains uncertain, melting experiments by McBirney (1975) showed that separation of a siliceous immiscible liquid may have produced late-stage granophyres in the Skaergaard intrusion.

In this paper we report on a thick tholeiitic North Mountain Basalt flow in Nova Scotia (Fig. 1) showing metre-scale layering with thin 'rhyo-

---

* Present address: Department of Geological Sciences, University of British Columbia, Okanagan College, 1000 K.L.O. Road, Kelowna, British Columbia, Canada V1Y 4X8.

Mineralogical Magazine, September 1992, Vol. 56, pp. 309–318

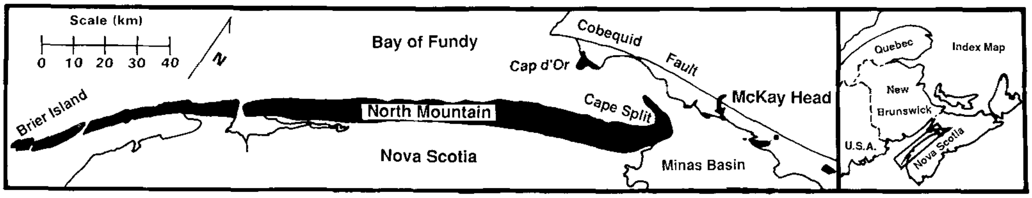

FIG. 1. Map showing the location of McKay Head and the distribution of North Mountain Basalt (solid black).

lite' bands produced by differentiation of the basalt. Much, though not all of the textural, mineralogical and chemical data suggest that the rhyolites formed through immiscibility, but the delicate, definitive evidence (globules of one glass inside another) may have been obscured by zeolite facies metamorphism.

Geology and field description

The North Mountain Basalt Formation (190 Ma; Hayatsu, 1979) consits of a thick (typically 150–200 m) lower flow and similarly thick upper flow separated by a 50 m thick middle unit of thin flows. The upper flow is overlain locally by 75 m of thin flows (Papezik *et al.*, 1988; Greenough *et al.*, 1989). The two thick flows are physically and isotopically traceable for more than 230 km along the Bay of Fundy (Fig. 1; Jones and Mossman, 1988; Greenough *et al.*, 1989; J. Colwell, pers. comm.). The basalts are chemically typical of high-Ti quartz-normative tholeiites from the Early Jurassic, Eastern North American dolerite province (Dostal and Dupuy, 1984; Papezik *et al.*, 1988).

Layering is apparent in the upper portions of the two thick flows at numerous localities around the Bay of Fundy but nowhere is it better displayed than in the ~175 m thick upper unit at KcKay Head (Fig. 1). The section at McKay Head dips to the west but the lowermost layered portion of the flow is not exposed due to a fault that cuts the section 34 m below the flow top. Layering is defined by ~25 cm thick mafic pegmatites which gradually change upward into similarly thick vesicular layers. Layers are separted from one another by ~130 cm thick sections of basalt (Fig. 2). From the flow top to the fault there is 5 m of unlayered basalt, 7 m of basalt with vesicular layers, and an 8 m layered transition zone followed by 14 m of basalt containing mafic pegmatite layers. These rocks underwent zeolite facies metamorphism (Aumento, 1966) but the pegmatites discussed here were relatively unaffected by metasomatism compared to vesicular rocks near the top of the flow (Greenough *et al.*, 1989).

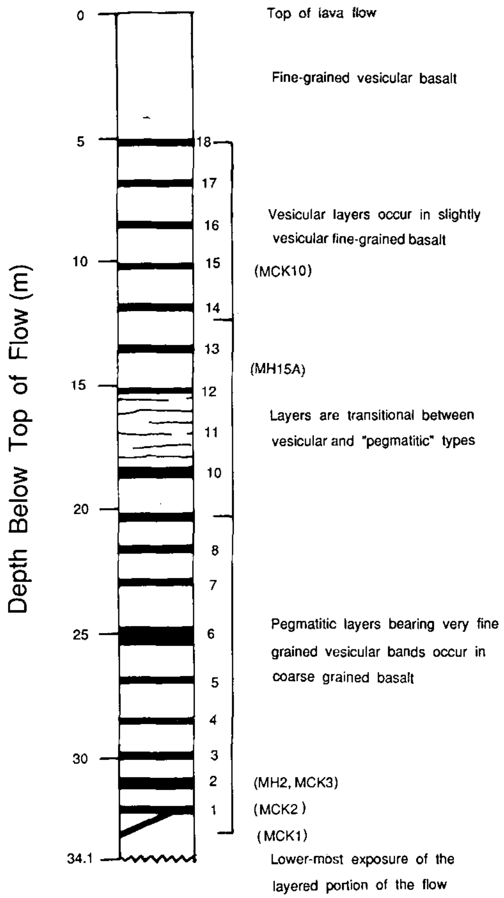

FIG. 2. Layering in the upper 35 m of the basalt flow at McKay Head. Broad dark lines are proportional in width to pegmatitic and vesicular differentiated layers. Depth in the flow (m) appears on the left. Layer numbers, sample locations and summary rock descriptions are given on the right. Diagram modified after Greenough and Dostal (in press.).

The various types of layering at McKay Head are described in detail in Greenough and Dostal (in prep.), but emphasis is placed here on the pegmatitic rocks. For descriptions of other mafic flows exhibiting layering, the reader is referred to Emerson (1905), Annells (1973), Helz (1980), and Puffer and Horter (1991).

The mafic pegmatite layers at McKay Head are 6 to 56 cm thick and show sharp to gradational contacts with interlayered, 80 to 180 cm thick, coarse-grained basalt. The lower pegmatitic layers locally display <15 cm long variolitic sheaths of augite which extend inward from the top and bottom of the layers. Typically, a fine-grained, sinuous, locally vesicular, rhyolite band 1–4 cm thick occurs in the middle of, and forms a sharp contact with each pegmatite layer (Fig. 3*a*). The bands are easily overlooked because they are similar in colour to the pegmatites, but their siliceous nature becomes apparent on petrographic or chemical analysis. In some of the thicker pegmatites, rhyolite occurs in crossing, discontinuous (<1 m long) stringers <1 cm wide. Vesicularity of the rhyolite bands tends to increase with height. Individual pegmatite layers and their rhyolite bands are traceable for 50 m along the shoreline.

Sampling and petrographic information

Sampling locations appear in Fig. 2. Mineral and mesostasis analyses reported in Table 1 and discussed in the text were made using wavelength-dispersive electron microprobes at The Memorial University of Newfoundland (JEOL JXA-50A) and the University of Saskatchewan (JEOL JXA-8600).

Basalt samples (MCK1, MH15A) from between layers display intergranular to subophitic textures with subequal proportions of tabular, zoned (An73–48) plagioclase (≤1.5 mm) and tabular to interstitial, commonly twinned, augite grains (≤2.0 mm) bearing fine pigeonitic exsolution lamellae locally. Analyses of augite show limited compositional variation and average

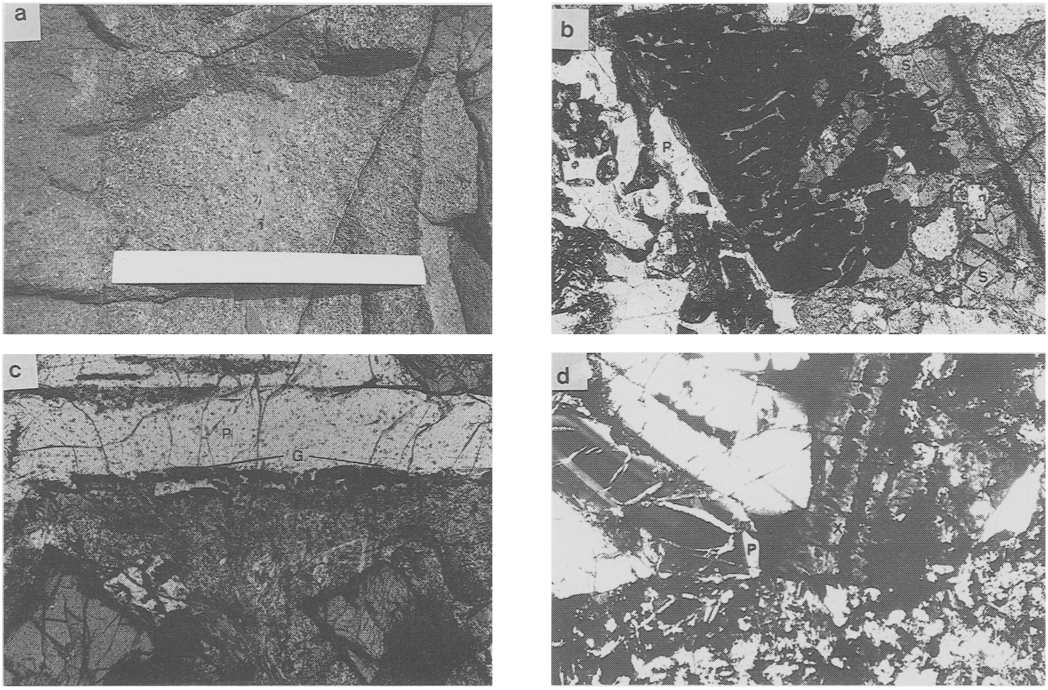

FIG. 3. (*a*) Pegmatite layer number 4 (Fig. 2) showing a narrow and vuggy (vesicular) rhyolite band in the middle. The upper and lower boundaries of the pegmatite occur near the ends of the 30 cm ruler. (*b*) Photomicrograph of a skeletal Fe-Ti-oxide grain in a rhomb-shaped mass of microcrystalline stilpnomelane (S) the latter bounded by augite on the far right (A) and plagioclase with poikilitic augite globules on the left (P) (plane polarised light, field width = 1.25 mm). (*c*) Pyroxene globules (G) along the edge of a plagioclase grain (P) and projecting locally into quartzo-feldspathic matrix (M) of a mafic pegmatite sample (plane polarised light, width of photograph = 3.25 mm). (*d*) Contact between a rhyolite band (lower portion of photograph) and mafic pegmatite. A long pyroxene crystal (X) and zoning in a plagioclase crystal (P) are truncated at the rhyolite band (crossed polars, field width = 3.25 mm).

Table 1 Analyses of minerals and mesostasis.

	Basalt		Rhyolite	Pegmatite						
	Plagioclase	Augite	Ferroaugite	Ferroaugite	Augite	Ferrorichterite	Stilpnomelane	Mesostasis	Ilmenite	Magnetite
	MCK1-3L	MH15A4X	MCK2F2X	MCK3B4X	MCK2B4X	MCK2B1A	MCK3T-S	MCK2BM	2B-I	2B-M
SiO_2	52.50	53.14	48.25	48.26	52.81	52.64	37.97	73.50	0.00	0.18
TiO_2	-	0.38	0.75	0.59	0.32	0.31	0.15	0.19	40.09	17.38
Al_2O_3	28.98	1.56	1.06	0.72	1.99	0.99	10.52	11.45	0.05	0.99
Cr_2O_3	-	0.15	0.00	0.02	0.27	0.00	-	-	-	-
FeO	0.89	9.17	26.41	30.54	8.36	19.99	18.82	0.70	48.43	74.31
MnO	-	0.20	0.54	0.69	0.18	0.08	0.12	0.03	0.36	0.39
MgO	-	16.98	4.25	6.59	17.60	7.98	12.92	0.36	0.17	0.09
CaO	12.51	18.05	17.89	11.84	18.65	6.45	1.76	0.25	0.02	0.00
Na_2O	4.31	0.14	0.16	0.09	0.28	6.70	0.37	3.09	0.04	0.00
K_2O	0.23	0.01	0.02	0.00	0.00	0.00	0.23	5.41	0.00	0.00
P_2O_5	-	-	-	-	-	-	0.02	0.03	0.00	0.03
TOTAL	99.42	99.63	99.33	99.34	100.19	95.14	82.88	100.01	89.16	93.37
Si	9.62	1.962	1.958	1.962	1.937	8.09	6.88		0.000	0.06
Al	6.26	0.068	0.051	0.034	0.086	0.18	2.25		0.003	0.40
Ti	-	0.012	0.026	0.020	0.010	0.04	0.02		1.867	5.00
Cr	-	0.004	0.000	0.001	0.008	0.00	-		-	-
Fe	0.14	0.283	0.896	1.038	0.256	2.57	2.86		2.226	21.10
Mn	-	0.006	0.019	0.024	0.006	0.01	0.02		0.017	0.11
Mg	-	0.934	0.257	0.399	0.962	1.83	3.49		0.014	0.05
Ca	2.46	0.714	0.778	0.516	0.733	1.06	0.34		0.001	0.00
Na	1.53	0.010	0.013	0.007	0.020	2.00	0.13		0.004	0.00
K	0.05	0.000	0.001	0.000	0.000	0.00	0.05		0.000	0.00
P							0.00		0.000	0.01

Notes: Major elements in oxide wt. % with total Fe as FeO. Mineral formulae calculated on basis of: Plagioclase and magnetite, O=32; clinopyroxene and ilmenite, O=6; amphibole, O=23; stilpnomelane, O=24. Mesostasis = average of 5 analyses with beam 10 microns wide.

Wo36 En44 Fs20. Minor Fe–Ti oxides form euhedral to skeletal grains in spatial association with interstitial intergrowths of quartz and feldspar in the mesostasis. Distinct pigeonite grains, pigeonitic exsolution lamellae in augite and pigeonite rimming augite show minor, local, preferential alteration to stilpnomelane (<15%). Alteration had little effect on plagioclase.

Pegmatite samples (MCK2B, 2T, 3B, 3T and MH2P) contain augite with a mean composition (Wo35 En41 Fs24) similar to that in the basalts, but grain edges are Fe-rich (Table 1). Locally, pyroxene is replaced by ferrorichterite (Table 1). In some samples (e.g. MCK2B and MCK2T), variolitic pyroxene needles cut large (4 mm), tabular, rarely needle-like, zoned (An62–41) plagioclase grains with the two minerals in approximately equal proportions. In other samples (e.g. MCK3B, 3T), pyroxenes occur as large (4 mm) tabular grains, as poikilitic and skeletal intergrowths in plagioclase grains (Fig. 3*b*), and as lobes on pyroxene grains, or as globules bordering plagioclase, adjacent to a quartzo-feldspathic matrix (Fig. 3*c*). Fe–Ti oxides typically show skeletal textures (Fig. 3*b*). Polygonal spaces (≤3 mm) between pyroxene and plagioclase grains contain quartz-needle and feldspar intergrowths (Fig. 3*c*) and/or graphic intergrowths, or fine-grained to microcrystalline tan coloured masses (devitrified glass) which comprise 20% of each sample and have highly siliceous compositions (Table 1, mesostasis). Edges of, and pigeonitic exsolution lamellae within, augite grains are locally altered to stilpnomelane. Most commonly, stilpnomelane forms interstitial polygonal, structureless, microcrystalline masses that resemble glass in that they extinguish under crossed polars. The Fe-rich stilpnomelane (Table 1) is spatially associated with (surrounds) large (≤2 mm) skeletal Fe–Ti oxide grains (Fig. 3*b*) and comprises 15–20% of each sample.

Zoning in pegmatite mineral grains is truncated at the rhyolite bands (Fig. 3*d*) and angular pieces of pegmatite occur locally within the band. The fine-grained (<0.5 mm) rhyolites (MCK2FG, 3FG, MH2FG) display a granular texture with plagioclase (An41–36) and potassium feldspar intergrown with needles of quartz. Accessory phases include anhedral Fe–Ti oxides (~2%) and pale green anhedral pyroxenes (~5%) with Fe- and Ca-rich compositions (Table 1). Minor (≤5%) polygonally shaped pockets of microcrystalline stilpnomelane occur between quartz needles and feldspar gains.

Geochemistry

Table 2 and Fig. 4 show that in comparison with the rhyolites and mafic pegamtites, the basalts have intermediate TiO_2 (1.4 wt.%, volatile free) and total Fe values (9.5 wt.% FeO) but the highest Al_2O_3, Cr, CaO and Sr concentrations. The mafic pegmatites have higher TiO_2 and FeO values (1.7 and 12.3 wt.% respectively) than the basalts or rhyolites. For many elements, concentrations in the basalts and pegmatites are similar. Some elements (e.g. MgO and P_2O_5) show large concentration variations in the pegmatites but the variations are not random. Complimentary pegmatite samples from above and below the rhyolite band are similar in composition (Fig. 4). The rhyolites display the lowest FeO, TiO_2, P_2O_5, and Sr concentrations and highest SiO_2, K_2O, Th and Zr.

The basalts and pegmatites have compositions closely resembling a Mesozoic tholeiite reported by Philpotts (1982) and known to contain immiscible Fe-rich and Si-rich glasses. Si-rich glass in the Connecticut tholeiite is chemically similar to the rhyolites (Table 2) except that P_2O_5 is slightly higher in the latter.

The geochemical effect of zeolite facies metamorphism is difficult to assess. Previous studies

Table 2 Averaged data, modelling results and comparative data.

	Average Basalt[1]	Average Pegmatite[2]	Average Rhyolite[3]	Model Rhyolite[4]	Connecticut Tholeiite[5]		
					Si-Glass	Fe-Glass	Rock
SiO_2	54.11	53.93	71.61	70.51	73.7	44.0	53.50
TiO_2	1.43	1.71	0.70	0.69	0.6	4.0	1.12
Al_2O_3	15.11	12.64	10.68	10.74	12.8	3.4	14.04
FeO	9.59	12.30	6.15	6.06	5.0	37.6	12.32
MnO	0.24	0.26	0.14	0.71	0.0	0.6	0.22
MgO	6.36	6.61	2.06	2.09	0.0	0.8	5.72
CaO	9.66	8.77	2.38	2.36	2.5	8.8	9.86
Na_2O	2.49	2.68	3.10	3.24	4.1	0.4	2.64
K_2O	0.86	0.92	2.82	2.95	1.3	0.1	0.34
P_2O_5	0.16	0.18	0.15	0.63	0.0	0.3	0.24
Rb	26	28	100				
Sr	197	182	124				
Ba	197	184	666				
Th	3.0	2.9	12				
Zr	124	125	415				
Y	27	26	46				
La	14	15	32				
Sm	4.4	4.9	9.0				
Eu	1.4	1.4	2.6				
Yb	2.4	2.9	4.1				
Sc	36	43	10				
V	303	375	29				
Cr	83	12	1				
Co	50	62	76				
Ni	26	22	1				
Ga	18	16	17				

Notes: Major elements in oxide wt. %, recalculated 100% volatile free.
Total Fe as FeO. Trace elements in ppm.
Average data from Greenough and Dostal (in prep.).
1. Average basalt = MCK1 & MH15A.
2. Average pegmatite = MCK2B, 2T, 3B and 3T.
3. Average rhyolite = MCK2FG and MCK3FG.
4. Rhyolite modelled using mass-balance calculations (Bryan et al., 1969) and mineral analyses in Table 1. Proportions of augite (MCK2B-4X):plagioclase (MCK13L):ilmenite:magnetite:stilpnomelane are 0.309:0.480:0.014:0.046:0.152. Percent crystallization = 75.1 and sum of squared residuals (R^2) = 1.8.
5. Fe-rich and Si-rich glass and a whole-rock analysis for tholeiitic basalt from Southbury Connecticut (Philpotts, 1982).

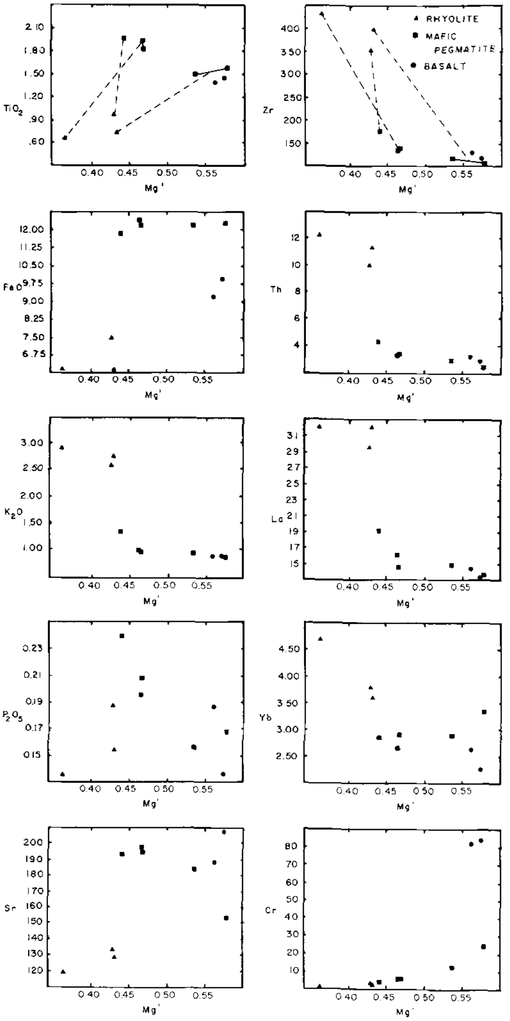

FIG. 4. Variation diagram showing selected major element oxides (wt.% volatile free, FeO = total Fe) and trace elements (ppm) plotted against the differentiation indicator Mg' [Mg' = Mg/(Mg + 0.9Fe) atomic]. Solid lines connnect pegmatite samples from above and below the rhyolite at the end of the dashed lines.

have shown that the alkali and alkaline earth metals were mobile during metamorphism of the North Mountain Basalt (Dostal and Dupuy, 1984; Papezik *et al.*, 1988; Greenough *et al.*, 1989). However, Sr isotopic studies show that carefully selected samples, such as those used here, yield geochemical data suitable for petrogenetic interpretation (Jones and Mossman, 1988). Thus the assumption is made that reported element concentrations primarily reflect original values.

Discussion

It is unlikely that the rhyolite bands represent a foreign siliceous magma because they occur at numerous locations along North Mountain as far as 225 km away from McKay Head. Further, it is difficult to propose a mechanism for the injection of 50+ m long by 2 cm wide rhyolite bands at regularly spaced intervals. Although sediments may have been assimilated at the base of the flow, it is doubtful that the bands represent partially assimilated material because of their height in the flow (near the top) and regular spacing. The bands must have formed from *in-situ* differentiation processes.

Polygonal interstices in the pegmatites contain quartzo-feldspathic intergrowths that are texturally identical to those in the rhyolite bands. Locally the bands are connected to pegmatite interstices along rhyolite-filled 'cracks'. Apparently the rhyolite liquid was derived from the pegmatites, possibly by fractional crystallisation or liquid immiscibility.

Crystal fractionation. Cumulus textures are absent in the flow, thus ruling out crystal settling, but crystal growth from the floor of the flow upward, or from the bottom and top of discrete layers, might explain the observed differentiation. This mechanism has been used to explain differentiation in many sills (Wager and Brown, 1967). Another possible crystal fractionation mechanism involves separation of residual interstitial liquid through compaction of a crystal mush or by gas-charged filter pressing. One problem with the crystal fractionation hypothesis is that it should have produced high P_2O_5 concentrations in the rhyolites (Anderson and Gottfried, 1971), which is not the case.

Mass balance calculations simulating crystal fractionation were incapable of reproducing rhyolite compositions from average basalt despite nearly 700 different mineral combinations being tried from a comprehensive and chemically diverse data set (70 analyses selected from over 300 available) of 5 *primary* basalt and pegmatite mineral phases. The smallest R^2 (sum of squared residuals) obtained for a reasonable set of minerals was ~30. In short, one or more phases necessary to effect mass balance is missing from the mineral data set. Adding stilpnomelane to the calculations produced much lower R^2 values (1.8; see model rhyolite, Table 2), suggesting that it represents the missing phase. Evaluation of the crystal fractionation hypothesis using trace element modelling techniques is difficult because modelled *REE* and *HFSE* concentrations are highly dependent on the percentage of apatite removed and this is difficult to assess. Nevertheless Sr and Ga decrease between basalt and rhyolite, an observation difficult to explain by crystal fractionation.

Liquid immiscibility. Globules of one liquid (glass) in another is the only definitive evidence for immiscibility. Perhaps as a result of devitrification, complete crystallisation or entire segregation, such evidence was not preserved in pegmatite samples. Nevertheless, some chemical and textural data strongly suggests that immiscibility occurred.

Textures indicative of immiscibility (Philpotts, 1978) during crystallisation of the pegmatites include: (1) globules of Fe-rich pyroxene (analysis MCK3B-4X, Table 1) with crystal faces growing into and partially surrounded by or surrounding quartzo-feldspathic intergrowths. These types of textures have been attributed to inhomogeneous nucleation of Fe-rich liquid globules on early pyroxene and plagioclase grains; (2) inclusions in plagioclase of fine-grained Fe-rich pyroxene and Fe–Ti oxides are consistent with iron-rich immiscible globules nucleating and locally coalescing on the surface of growing plagioclase crystals. Later the globules crystallised as pyroxene, oxides and a rim of zoned Na-rich plagioclase in optical continuity with the original plagioclase.

Both Fe- and Si-rich glass may have originally existed in the pegmatites. Locally, the siliceous material filling polygonal interstices is microcrystalline and resembles devitrified glass. Other polygonal interstices contain internally structureless microcrystalline stilpnomelane which surrounds, partially or totally, large skeletal ilmenite-rich oxide grains (Fig. 3*b*). A large percentage of the stilpnomelane (70%) shows these textural relationships with oxide grains, although there is the possibility that some of the mineral represents gas cavity infilling. It seems probable that the Fe–Ti oxide grains grew from a supercooled Fe-rich liquid (hence the skeletal texture; Lofgren, 1974) and that the last portions of the liquid were quenched to form glass and later altered to stilpnomelane. The stilpnomelane chemically resembles some immiscible Fe-rich glasses though TiO_2 and P_2O_5 are low and MgO high (compare Table 1, stilpnomelane and Fe-glass, Table 2). The low TiO_2 concentrations may reflect growth of the Ti-rich oxide grains prior to solidification of an Fe-rich liquid. The stilpnomelane appears sensitive to local MgO diffusion because analyses from other pegmatite localities around the Bay of Fundy show higher FeO (total Fe) and lower MgO contents (36 and 2 wt.%, respectively) which are even closer to the Fe-rich glass composition. Phosphorus may have been redistributed in a similar fashion and concentrated in apatite grains.

The degree of crystallisation required for the onset of immiscibility, and typical relative volumes of the two liquids are known from studies of basaltic rocks (e.g. Roedder and Weiblen, 1971; Philpotts, 1982). These percentages and proportions can be estimated by treating the pegmatite–rhyolite layer as a whole, and adding the modal percentages of pegmatite quartzo-feldspathic matrix, rhyolite (in bands), and interstitial stilpnomelane in the pegmatites. These calculations indicate that by, or prior to 59 volume percent crystallisation of an initial liquid with combined pegmatite + rhyolite composition (approximately basaltic), two immiscible liquids had formed. The volume proportions of Fe-rich liquid to Si-rich liquid are 32:68. Thus both the total percentage of immiscible liquids (41 vol.%) and relative proportions of Fe-rich to Si-rich liquid are typical of values recorded for basaltic rocks (e.g. Roedder and Weiblen, 1971; Philpotts, 1982).

Plagioclase and augite coexisting with immiscible globules in tholeiitic rocks typically have compositions of An50 and Wo34En19Fs47 respectively (Philpotts, 1982). Pegmatite plagioclase (An60–An41) and augite (Wo39En46Fs15–Wo27En234Fs50) compositions encompass or approach these values. The pyroxenes coexist with minor pigeonite and give temperatures ranging from 1150 °C to below 980 °C using the Davidson and Lindsley (1985) two-pyroxene geothermometer. These temperatures overlap typical experimentally determined values for the onset of immiscibility in tholeiites of just over 1000 °C (Philpotts and Doyle, 1983).

Fig. 5 illustrates that rhyolite compositions are consistent with immiscibility and that the pegmatites are comparable to basalts containing immiscible Fe- and Si-rich liquids (Philpotts, 1982). These rocks are similar to other Eastern North American tholeiites bearing immiscible liquids (compare basalt, pegmatite and rhyolite with Connecticut tholeiite analyses, Table 2). Of particular significance is their high total Fe content important to effecting immiscibility.

Philpotts (1982) argued that two factors which help to determine the quantity of liquid remaining when immiscibility occurs (i.e. the percentage of liquid that will segregate to form two liquids) are the degree of magma evolution and oxidation potential. The average value of the differentiation indicator Mg' (where Mg' = Mg/(Mg + 0.9 × total Fe) atomic) for pegmatite samples (~0.52) falls in the middle of the range for subalkaline basaltic rocks hosting immiscible liquids (Phil-

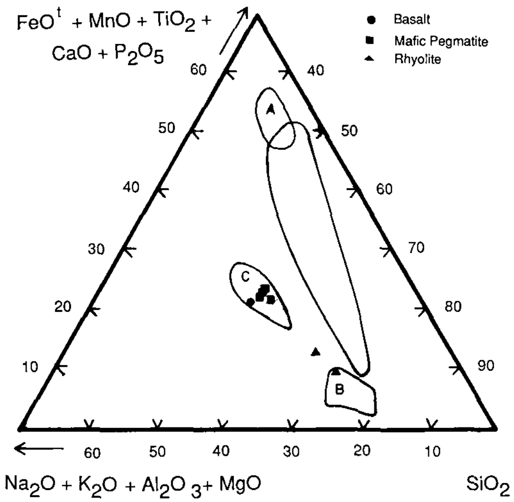

FIG. 5. Plot of McKay Head rocks on the immiscibility diagram of Philpotts (1982) as modified from Greig (1927). Circled fields labelled A, B and C are for immiscible iron-rich glasses and silica rich glasses and their tholeiitic host rocks (Philpotts, 1982). Also shown is the immiscibility field for the system fayalite–leucite–silica from Roedder (1951).

potts, 1982; 0.61–0.30). Early crystallisation of magnetite (as equant non-dendritic crystals) under oxidising conditions depletes the melt in iron and diminishes the amount of immiscible Fe-rich liquid that can form. Skeletal oxide grains (late-forming and indicative of quenching; Lofgren, 1974) in McKay Head rocks suggest low f_{O_2} conditions suitable for significant immiscibility.

Natural rocks show an immiscibility field similar to that in the synthetic system fayalite–leucite–silica (Roedder, 1951), except that titanium and phosphorus (and lower temperatures of formation) in the former tend to expand the immiscibility field (Freestone, 1978; Visser and Loster van Groos, 1979). Pegmatite P_2O_5 (0.19, wt.%, volatile free) and TiO_2 concentrations (1.7) fall at the lower end of, and in the middle of (respectively), the range in basaltic rocks showing immiscible liquids (Philpotts, 1982). The lower P_2O_5 content of the rhyolites compared to the pegmatites (Table 2) also supports immiscibility because partitioning of P_2O_5 into an Fe-rich liquid results in immiscibility-related rhyolites with low P_2O_5 concentrations (Roedder, 1978).

The immiscibility hypothesis can be evaluated given Fe-rich/Si-rich liquid partitioning coefficients for various trace elements. Assuming that immiscibility occurred, there is no obvious method to quantitatively determine the composition of the Fe-rich liquid, but comparison of relative trace element concentrations in the basalts, pegmatites, and rhyolites (Table 2) gives qualitative information on element behaviour. For example, the average Zr concentration in the pegmatites is the same as in the basalts but $\frac{1}{3}$ of that in the rhyolites. Thus the Fe-rich liquid retained in the pegmatites probably had a low Zr content. This analysis indicates that Rb, Ba, Th, and Zr partitioned into the Si-rich melt and Sr and V entered the Fe-rich melt (or Fe–Ti oxides controlled V). Chromium, Ni and Sc partitioned into the Fe-rich melt or were controlled by pyroxene. Whether most *REE*, Y, Nb, and Ga preferred the Fe-rich or Si-rich liquid cannot be determined.

In synthetic system melting experiments (Watson, 1976; Ryerson and Hess, 1978), high charge density cations (P, Ti, Zr, Ta, Cr, and the *REE*) were strongly enriched in the Fe-rich liquid, the alkaline earth metals (Sr, Ba) showed slight to negligible enrichment in the Fe-rich liquid, and the alkali metals (K, Rb, Cs) were strongly enriched in the Si-rich liquid. The antithetic behaviour of Th, Zr, Hf and Ba relative to the experimental results cannot be explained through immiscibility, suggesting that the effects of other processes may be superimposed.

One possible explanation for the antithetic behaviour of Th, Zr, Hf and Ba is that the movement of magmatic volatiles syn- or post-crystallisation altered the distribution of some elements between the pegmatites and rhyolites. Amphibole replacing primary augite locally in the pegmatites (Table 1) is not characteristic of zeolite facies metamorphism and suggests that deuteric alteration occurred. Considering the evidence for immiscibility and limited ability of deuteric processes to produce large-scale redistribution of the major elements, it is suggested that if dueteric processes affected the rocks it is more likely that they overprint the effects of immiscibility than fractional crystallisation.

A second possible reason for the antithetic behaviour of Th, Zr, Hf and Ba is that the volatile-free synthetic-system experiments are not entirely applicable to some natural systems. Hypothetically, volatiles (water) could depolymerise the siliceous liquid, thus modifying the distribution of elements between immiscible fluids compared to the experiments.

Mechanisms for rhyolite segregation. If formed through immiscibility, the rhyolite liquid would have been 4 orders of magnitude more viscous than the Fe-rich liquid (see viscosity estimates in Philpotts and Doyle, 1980, and Philpotts, 1982). This poses the question of why the rhyolite separated from the pegmatite and not the Fe-rich liquid? The occurrence of minor polygonal-shaped masses of stilpnomelane in the rhyolites plus high P_2O_5 concentrations (compared with the Connecticut Si-rich glass, Table 2) in the rhyolites indicate that some portion of the Fe-rich liquid did separate with the Si-rich glass. This aside, broken pegmatite crystals at the rhyolite interface show that pegmatite crystallisation occurred largely prior to formation of the rhyolite band. Vesicles in the rhyolite suggest that 'filter pressing' (Propach, 1976; Anderson *et al.*, 1984), involving gas exsolution from interstitial magma, drove the rhyolite liquid into low pressure fractures forming the rhyolite bands. Given this mechanism analogy with petroleum reservoirs may explain why the Fe-rich liquid was largely not removed. Oil–water movement in petroleum reservoirs demonstrates that viscosity has a limited influence on the relative permeability of a porous medium to two immiscible liquids (Drake, 1978, pp. 103–124). Generally, the liquid with smallest volume (i.e. water in oil reservoirs and the Fe-rich liquid in the pegmatites) and greatest mineral–surface wetting capacity will have the least ability to move through the porous medium. The liquid with the greater wetting capacity is difficult to determine because calculations presented in Philpotts (1990, p. 231) indicate that the Si-rich liquid would wet feldspar grains whereas the Fe-rich liquid would wet pyroxene grains. However, the pegmatite should be more permeable to the siliceous liquid due to its greater volume. Thus the rhyolite composition of the band is no more consistent with fractional crystallisation than with liquid immiscibility.

Synopsis and summary

The evidence for immiscibility may be summarised as follows: (1) textures such as fine-grained Fe-rich pyroxene globules on the surface of feldspar crystals argue for an Fe-rich liquid. (2) structureless, microcrystalline, interstitial, polygonal patches of Si-rich minerals and similar patches of stilpnomelane associated with skeletal Fe–Ti oxide grains have bulk chemical compositions (to a first approximation), relative proportions and total modal percentages generally consistent with their once having been Si-rich and Fe-rich glasses. (3) The basalts and pegmatites have bulk-rock compositions (particularly Fe, Ti and P), similar to rocks containing documented examples of immiscibility. (4) The major element composition of the rhyolite bands is consistent with their largely representing a siliceous liquid. (5) Pegmatite mineral compositions and inferred temperatures of formation are both compatible with immiscibility. The antithetic behaviour of Th, Zr, Hf and Ba suggests that late-stage volatiles redistributed these elements or that volatiles depolymerised the siliceous liquid and modified trace element partitioning between the immiscible liquids. Mass balance calculations show that the alternative to immiscibility—fractional crystallisation—is incapable of explaining the major element composition of the rhyolite bands given primary mineral phases in the basalts and pegmatites. The 'missing' phase is represented by stilpnomelane. Stilpnomelane commonly occurs in 'granophyres' from the Skaergaard complex, Greenland (Deer *et al.*, 1966). McBirney (1975) reported that at least some of these granophyres display an immiscible relationship with adjacent Fe-rich mafic rocks.

We suggest that the evidence for immiscibility forming the rhyolite bands is strong enough that studies on other thick flows are warranted. The rhyolite bands apparently represent the incipient stages of granophyre formation in intrusions. If granophyres form through immiscibility then rhyolite bands in thick flows appear suited to 'bridging the gap' texturally and chemically, between totally segregated granophyres and immiscible globules in thin basalt flows.

Acknowledgements

Mr. K. Cameron carried out chemical analyses and Ms D. Fan and M. Halko did the art work. Discussions with Drs A. Philpotts, G. Pajari, J. Colwell, J. Hodych, M. Reeves, and R. Kerrich are gratefully acknowledged. The manuscript benefited from comments by an anonymous referee. This research was funded by NSERC grants to the authors.

References

Anderson, A. T. and Gottfried, D. (1971) Contrasting behaviour of P, Ti and Nb in a differentiated high-alumina olivine–tholeiite and a calc-alkaline andesite suite. *Geol. Soc. Amer. Bull.*, **82**, 1929–42.

Anderson, A. T., Swinhart, G. H., Artioli, G., and Geiger, C. A. (1984) Segregation vesicles, gas filter-pressing, and igneous differentiation. *J. Geol.*, **92**, 55–72.

Annells, R. N. (1973) Proterozoic flood basalts of eastern Lake Superior. The Keweenawan volcanic rocks of the Mamainse Point area, Ontario. *Geol. Surv. Can.*, Paper 72–10, 51pp.

Aumento, F., (1966) Zeolite minerals, Nova Scotia. In *Geology of Parts of the Atlantic Provinces*. Geological Association of Canada, Mineralogical Association of Canada Guidebook, 71–7.

Bowen, N.L. (1928) *The Evolution of the Igneous Rocks*. Princeton University Press, Princeton, N.J.

Bryan, W. B., Finger, L. W., and Chayes, F. (1969)

Estimating proportions in petrographic mixing equations by least-squares approximation. *Science,* **163**, 926–7.

Davidson, P. M. and Lindsley, D. H. (1985) Thermodynamic analysis of quadrilateral pyroxenes Part II, Model calibration from experiments and applications to geothermometry. *Contrib. Mineral. Petrol.,* **91**, 390–404.

De, A. (1974) Silicate liquid immiscibility in the Deccan traps and its petrogenetic significance. *Geol. Soc. Amer. Bull.,* **85**, 471–4.

Deer, W. A., Howie, R. A. and Zussman, J. (1966) *An Introduction to the Rock-Forming Minerals.* Longman, London, 528pp.

Dostal, J. and Dupuy, C. (1984) Geochemistry of the North Mountain basalt (Nova Scotia, Canada). *Chem. Geol.,* **45**, 245–61.

Drake, L. P. (1978) *Fundamentals of Reservoir Engineering.* Elsevier Scientific Publishing Company, Amsterdam, 433pp.

Emerson, B. K. (1905) Plumose diabase and palagonite from the Holyoka trap sheet. *Geol. Soc. Amer. Bull.,* **16**, 91–130.

3Freestone, I. C. (1978) Liquid immiscibility in alkali-rich magmas. *Chem. Geol.,* **23**, 115–23.

Greenough, J. D. and Dostal, J. (in press.) Cooling history and differentiation of a thick North Mountain Basalt flow (Nova Scotia, Canada). *Bull. Volcan.*

——Jones, L. M. and Mossman, D. (1989) Petrochemical and stratigraphic aspects of North Mountain basalt from the north shore of the Bay of Fundy, Nova Scotia, Canada. *Can. J. Earth Sci.,* **26**, 1701–7.

Greig, J. W. (1927) Immiscibility in silicate melts. *Amer. J. Sci.,* ser. 4, **13**, 1–44 & 133–54.

Hayatsu, A. (1979) K–Ar isochron age of the North Mountain basalt, Nova Scotia. *Can. J. Earth Sci.,* **16**, 973–5.

Helz, R. T. (1980) Crystallisation history of Kilauea Iki lava lake as seen in drill core recovered in 1967–1979. *Bull. Volcan.,* **43**, 675–701.

Jones, L. M., and Mossman, D. J. (1988) The isotopic composition of strontium and the source of the Early Jurassic North Mountain basalt, Nova Scotia. *Can. J. Earth Sci.,* **25**, 942–4.

Lofgren, G. (1974) An experimental study of plagioclase crystal morphology: isothermal crystallisation. *Amer. J. Sci.,* **274**, 243–73.

McBirney, A. R. (1975) Differentiation of the Skaergaard intrusion. *Nature,* **253**, 691–3.

Papezik, V. S., Greenough, J. D., Colwell, J., and Mallinson, T. (1988) North Mountain basalt from Digby, Nova Scotia: Models for a fissure eruption from stratigraphy and petrochemistry. *Can. J. Earth Sci.,* **25**, 74–83.

Philpotts, A. R. (1976) Silicate liquid immiscibility: its probable extent and petrogenetic significance. *Amer. J. Science,* **276**, 1147–77.

——(1978) Textural evidence for immiscibility in tholeiite. *Mineral. Mag.,* **42**, 417–25.

——(1979) Silicate liquid immiscibility in tholeiite basalts. *J. Petrol.,* **20**, 99–118.

——(1982) Compositions of immiscible liquids in volcanic rocks. *Contrib. Mineral. Petrol.,* **80**, 201–18.

——(1990) *Principles of Igneous and Metamorphic Petrology.* Prentice Hall, Englewood Cliffs, New Jersey, 498pp.

——and Doyle, C. D. (1980) Immiscibility in tholeiites: a discussion. *Mineral. Mag.,* **43**, 939–40.

————(1983) Effect of magma oxidation state on the extent of silicate liquid immiscibility in a tholeiite basalt. *Amer. J. Sci.,* **283**, 967–86.

Propach, G. (1976) Models of filter differentiation. *Lithos,* **9**, 203–9.

Puffer, J. H. and Horter, D. L. (1991) Basaltic pegmatites of the Lolo Basalt, Washington and the Watchung Basalts, New Jersey. *Northeastern Section, Geological Society of America Abstracts with Programs,* **23**, 118.

Roedder, E. (1951) Low temperature liquid immiscibility in the system K_2O–FeO–Al_2O_3–SiO_2. *Amer. Mineral.* **36**, 282–6.

——(1978) Silicate liquid immiscibility in magmas and in the system K_2–FeO–Al_2O_3–SiO_2: an example of serendipity. *Geochem. Cosmochim. Acta,* **42**, 1597–617.

——and Weiblen, P. W. (1970) Lunar petrology of silicate melt inclusions, Apollo 11 rocks. In Apollo 11 Lunar Science Conference Proceedings, *Geochim. Cosmochim. Acta,* Supplement 1, **1**, 801–37.

————(1971) Petrology of silicate melt inclusions, Apollo 11 and Apollo 12 and terrestrial equivalents. Proceedings of the Second Lunar Science Conference. Ibid., Supplement 2, **1**, 507–28.

Ryerson, F. J. and Hess, P. C. (1978) Implications of liquid–liquid distribution coefficients to mineral–liquid partitioning. *Geochim. Cosmoch. Acta,* **42**, 921–32.

Visser, W. and Koster van Groos, A. F. (1979) Effects of P_2O_5 and TiO_2 on liquid–liquid equilibria in the system K_2O–FeO–Al_2O_3–SiO_2. *Amer. J. Sci.,* **279**, 970–89.

Wager, L. R. and Brown, G. M. (1967) *Layered Igneous Rocks.* Oliver and Boyd, London, 588pp.

Watson, E. B. (1976) Two-liquid partition coefficients: Experimental data and geochemical implications. *Contrib. Mineral. Petrol.* **56**, 119–34.

[*Manuscript received 27 July 1991: revised October 20 1991*].